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THERMAL EXPANSION OF  $\text{KYb}_x\text{Y}_{1-x}(\text{WO}_4)_2$  CRYSTALS

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**Abstract.** Average and differential linear thermal expansion coefficients of  $\text{KYb}_x\text{Y}_{1-x}(\text{WO}_4)_2$  crystals are determined in the directions of the optical indicatrix axes  $N_p$ ,  $N_m$  and  $N_g$  using dilatometric technique. Dependence of the average and differential linear thermal expansion coefficients on the temperature in the range of 75–290 °C and ytterbium contains  $x$  from 0 to 1.0 are discussed.

**Key words:** monoclinic crystals, linear thermal expansion coefficients,  $\text{KYb}_x\text{Y}_{1-x}(\text{WO}_4)_2$  crystals.

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ТЕРМИЧЕСКОЕ РАСШИРЕНИЕ КРИСТАЛЛА  $\text{KYb}_x\text{Y}_{1-x}(\text{WO}_4)_2$

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**Аннотация.** Определены средние и дифференциальные термические коэффициенты линейного расширения кристаллов  $\text{KYb}_x\text{Y}_{1-x}(\text{WO}_4)_2$  в направлениях осей оптической индикатрисы  $N_p$ ,  $N_m$  и  $N_g$  с помощью dilatометрического метода. Обсуждаются зависимости данных коэффициентов от температуры в диапазоне 75–290 °C и содержания иттербия  $x$  от 0 до 1,0.

**Ключевые слова:** моноклинные кристаллы, термические коэффициенты линейного расширения, кристаллы  $\text{KYb}_x\text{Y}_{1-x}(\text{WO}_4)_2$ .

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Potassium-yttrium tungstate crystals doped with ytterbium ions [ $\text{KYb}_x\text{Y}_{1-x}(\text{WO}_4)_2$ ] belong to the most promising laser materials for generating ultrashort laser pulses of a high average power. The advantages of such media are high absorption and stimulated emission cross sections combined with relatively wide gain bands and intermediate thermal conductivity. Moreover, the Stokes shift between the pump and lasing wavelengths (laser quantum defect) for these Yb-doped crystals is very small, ~ 5 %, which determines the low heat dissipation in an laser active medium. These properties of  $\text{KYb}_x\text{Y}_{1-x}(\text{WO}_4)_2$  crystals make it possible to obtain high generation efficiencies in different lasing modes (cw, Q-switching, mode locking, regenerative amplification, planar waveguide lasers). The ionic radii of  $\text{Y}^{3+}$  and  $\text{Yb}^{3+}$  are similar (1.019 Å and 0.985 Å, respectively), and the  $\text{KY}(\text{WO}_4)_2$  crystal gives the possibility of continuous Yb-doping reaching the  $\text{KYb}(\text{WO}_4)_2$  one.

A knowledge of the thermal expansion properties of laser crystals is of great engineering importance, particularly, to estimate thermo-optic coefficient (refractive index change due to temperature change) which is one of the most important parameters governing thermooptical distortions of the laser medium. However, the

thermal expansion properties of the  $\text{Yb:KY}(\text{WO}_4)_2$  at different Yb-doping level has not been studied in detail to date. In this report, we present the measurements of linear thermal expansion coefficients (LTEC) of  $\text{KYb}_x\text{Y}_{1-x}(\text{WO}_4)_2$  ( $\text{KYb}_x\text{Y}_{1-x}\text{W}$ ) crystals with ytterbium concentration from 0 to 100 at.% ( $x = 0-1.0$ ).

The growth of  $\text{KYb}_x\text{Y}_{1-x}\text{W}$  crystals was carried out by the modified Czochralski method using the dynamic growth regime and the selection of the required temperature gradient at the crystallization front. The  $\text{KYb}_x\text{Y}_{1-x}\text{W}$  are monoclinic crystals with space group  $C2/c$ , and thus they are optically biaxial ones. Low symmetry of the  $\text{KYb}_x\text{Y}_{1-x}(\text{WO}_4)_2$  lattice results in significant anisotropy of its optical properties. Optical properties of the  $\text{KYb}_x\text{Y}_{1-x}\text{W}$  crystals are described within the frame of the optical indicatrix with orthogonal principal axes  $N_p$ ,  $N_m$ , and  $N_g$ . Axis corresponding to the minimum principal refractive index ( $N_p$ ), coincides with the crystallographic axis  $b$ , while two others ( $N_m$  and  $N_g$ ) are positioned in the  $a-c$  plane [1, 2].

To characterize the anisotropy of the LTECs in  $\text{KYb}_x\text{Y}_{1-x}\text{W}$  crystals, one sample from each crystal composition (ytterbium content,  $x$ ) was prepared in a shape of a rectangular parallelepiped along the  $N_p$ ,  $N_m$ , and  $N_g$  directions, respectively.

The LTECs  $\alpha_p$ ,  $\alpha_m$ , and  $\alpha_g$  (along the axes  $N_p$ ,  $N_m$ , and  $N_g$ , respectively) were measured by means of Horizontal pushrod dilatometer Netzsch DIL 402 PC. The measurements were performed over the temperature range 25 °C to 290 °C This temperature range is of interest when the crystals under study are used as active laser media. The error in determining of the LTECs was  $\sim 0.3 \times 10^{-6} \text{ K}^{-1}$ . The values of the LTECs along the principal optical axis are more important in practice because normally the laser elements are cut along one of these axes.

The average LTEC is governed by the slope of the chord between two points on the curve describing dependence of the length on temperature. According to this, the average LTECs along a given direction were calculated for different temperature ranges ( $T-T_0$ ) using:  $\bar{\alpha} = \frac{1}{L_0} \frac{(L_T - L_0)}{(T - T_0)}$ , where  $L_0$  and  $L_T$  are the sample lengths at initial (room)  $T_0 = 25 \text{ °C}$  and terminal  $T$  temperatures of the range, respectively. The typical dependence of average LTECs  $\bar{\alpha}$  on the terminal temperature  $T$  along the principal optical axis  $N_p$ ,  $N_m$ , and  $N_g$  is shown in Fig. 1(a) for ytterbium content  $x = 0.05$ . The anisotropy of the average LTEC is characterized by  $\bar{\alpha}_g > \bar{\alpha}_m > \bar{\alpha}_p$ , which is consistent with the [3] data for KYW and KYbW. The  $\bar{\alpha}$  values increase with increasing temperature range which is usually observed for oxide crystals (in particular, [4, 5]). The values of average LTECs  $\bar{\alpha}$  for each direction as a function of ytterbium content  $x$  at temperature 200 °C are presented in Fig. 1 (b). It is seen that an increase in the ytterbium content  $x$  leads to a decrease in the  $\bar{\alpha}_g$  value. This decreasing can be approximated by a linear dependence  $\bar{\alpha}_g = (16.4 - 1.17x) 10^{-6} \text{ K}^{-1}$ , the linear approximation gives a fairly high rate of the coefficient of determination  $R^2 = 0.86$ . The difference between the  $\bar{\alpha}_g$  values for  $x = 0$  (KYW) and  $x = 1.0$  (KYbW) is  $\sim 1.2 \times 10^{-6} \text{ K}^{-1}$ , which is four times greater than the LTEC determining accuracy ( $\sim 0.3 \times 10^{-6} \text{ K}^{-1}$ ). For the average LTEC along the  $N_p$  direction, doping of the KYW crystal with Yb ions (increasing of  $x$ ) does not results in change in the value of  $\bar{\alpha}_p$  (within the experimental error). The linear fitting of this dependence gives the coefficient of determination close to zero ( $R^2 = 0.0077$ ). Increasing the ytterbium content  $x$  leads to an increase in the value of  $\bar{\alpha}_m$ , which can be described by a linear relationship dependence  $\bar{\alpha}_m = (12.0 - 0.75x) 10^{-6} \text{ K}^{-1}$  with the coefficient of determination  $R^2 = 0.63$ . The relatively low coefficient  $R^2$  can be explained by a weak dependence of  $\bar{\alpha}_m$  on  $x$ . The difference between the  $\bar{\alpha}_m$  values for  $x = 0$  and  $x = 1.0$  is  $\sim 0.75 \cdot 10^{-6} \text{ K}^{-1}$ , which is only 2.5 times greater than the LTEC measurement error ( $\sim 0.3 \cdot 10^{-6} \text{ K}^{-1}$ ).

There are previous data on LTECs for KYW ( $x = 0$ ) and KYbW ( $x = 1.0$ ) measured along the  $N_p$ ,  $N_m$  and  $N_g$  axes which were obtained using dilatometric technique [6] as well the X-ray powder diffraction

analysis [3,7] (the unit cell parameters as a function of temperature were measured for crystallographic directions  $a$ ,  $b = N_p$ ,  $c$ , and  $c^* [c^* \cdot L_a]$ , the values along the  $N_g$  and  $N_m$  directions were calculated). The  $\bar{\alpha}_g$ ,  $\bar{\alpha}_m$ ,  $\bar{\alpha}_p$  values were reported in the range of (15.9–15.99), (10.3– 10.31), (1.9–2.0) for KYW and (13.7–14.47), (10.83–11.4), (2.0–2.6) for KYbW [ $10^{-6} \text{ K}^{-1}$ ], respectively [3, 6, 7]. The average LTEC values obtained in present paper are falling fairly well within the published range. It should be noted that the LTEC values in [3–7] were evaluated from the slopes of the measured expansion curves, and the dependence of  $\bar{\alpha}$  on temperature was not analyzed.

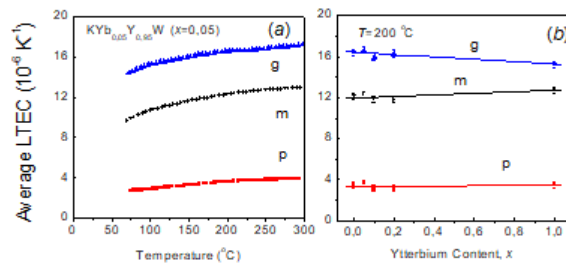


Figure 1 – Average LTEC  $\bar{\alpha}$  along the principal optical axis  $N_p$ ,  $N_m$ , and  $N_g$  as a function of terminal temperature  $T$  (a) and versus ytterbium content  $x$  (b) for  $\text{KYb}_x\text{Y}_{1-x}\text{W}$  crystals. Symbols – experimental data, lines in (b) – the linear approximations

In Fig. 2 (a) is shown typical differential LTECs  $\alpha$  for  $\text{KYb}_x\text{Y}_{1-x}\text{W}$  crystals versus temperatures along the  $N_p$ ,  $N_m$ , and  $N_g$  axis for ytterbium content  $x = 0.05$ .

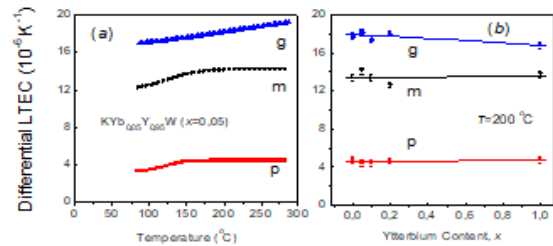


Figure 2 – Differential LTEC  $\bar{\alpha}$  along the principal optical axis  $N_p$ ,  $N_m$ ,  $N_g$  as a function of temperature  $T$  (a) and versus ytterbium content  $x$  (b) for  $\text{KYb}_x\text{Y}_{1-x}\text{W}$  crystals. Symbols – experimental data, lines in (b) – the linear approximations

The differential LTEC coefficient is related to the slope of the tangent of the length  $L$  versus temperature  $T$  plot. The  $\alpha$  value for different temperatures thus was obtained using the first derivative of the length with respect to the temperature as  $\alpha = \frac{1}{L_T} \left( \frac{dL}{dT} \right)_T$ , where  $L_T$  and  $(dL/dT)_T$  are the sample length and its temperature derivative at a given temperature  $T$ , respectively. Because  $\bar{\alpha}$  is not constant over the temperature range then  $\bar{\alpha} \neq \alpha$ . For the same temperature  $T$ , the  $\alpha$  values are slightly larger than the corresponding  $\bar{\alpha}$  ones. The anisotropy of the differential LTEC, like the average ones,

is characterized by the ratio  $\alpha_g > \alpha_m > \alpha_p$ . With increasing temperature, the value of  $\alpha$  increases, but at temperatures exceeding  $\sim 180$  °C it remains practically unchanged. The  $\alpha$  values for each direction as a function of ytterbium content  $x$  at temperature 200 °C are shown in Fig. 2 (b). An increase in the ytterbium content  $x$  leads to an decrease in the  $\alpha_g$  value which can be approximated by a linear dependence  $\alpha_g = (18.0 - 1.12x) 10^{-6} K^{-1}$ , the linear approximation gives a fairly high rate of the coefficient of determination  $R^2 = 0.70$ . The difference between the  $\alpha_g$  values for  $x = 0$  (KYW) and  $x = 1.0$  (KYbW) is  $\sim 1.1 \cdot 10^{-6} K^{-1}$ , which is about four times large as the uncertainty in the  $\alpha$  determination ( $\sim 0.3 \cdot 10^{-6} K^{-1}$ ). Along the  $N_m$  and  $N_p$  directions, doping with the Yb ions (increasing of  $x$ ) does not results in change in the values of  $\alpha_m$  and  $\alpha_p$  (within the experimental error). The linear fitting of this dependences gives low values of  $R^2$  (0.032 and 0.24, respectively). Moreover, the difference between the LTECs values for  $x = 0$  and  $x = 1.0$  ( $\sim 0.2 \cdot 10^{-6} K^{-1}$ ) is less than the LTEC measurement error ( $\sim 0.3 \cdot 10^{-6} K^{-1}$ ).

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#### HERMO-OPTIC COEFFICIENTS OF MONOCLINIC Er<sup>3+</sup>:(GdY)<sub>2</sub>SiO<sub>5</sub> CRYSTAL

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**Abstract.** Thermo-optic coefficients of the Er<sup>3+</sup>-doped gadolinium-yttrium oxyorthosilicate crystal Er<sup>3+</sup>:GdYSO are determined at a wavelength of 632.8 nm for light polarizations  $E // N_p, N_m$  and  $N_g$ . Linear thermal expansion coefficients are estimated for this crystal in the directions of the optical indicatrix axes  $N_m$  and  $N_g$ .

**Key words:** monoclinic crystal, thermo-optic coefficient, (GdY)<sub>2</sub>SiO<sub>5</sub> crystal, thermal coefficients of the optical path.

#### ТЕРМООПТИЧЕСКИЕ КОЭФФИЦИЕНТЫ МОНОКЛИННОГО КРИСТАЛЛА Er<sup>3+</sup>:(GdY)<sub>2</sub>SiO<sub>5</sub>

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**Аннотация.** Определены термооптические коэффициенты кристалла оксиортоосиликата гадолиния-иттрия, легированного ионами Er<sup>3+</sup> (Er<sup>3+</sup>:GdYSO, на длине волны 632,8 нм для поляризаций света  $E // N_p, N_m$  и  $N_g$ . Произведена оценка термических коэффициентов линейного расширения данного кристалла в направлениях осей оптической индикатрисы  $N_m$  и  $N_g$ .

**Ключевые слова:** моноклинный кристалл, термооптический коэффициент, кристалл (GdY)<sub>2</sub>SiO<sub>5</sub>, термический коэффициент оптического пути.

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Yttrium and gadolinium oxyorthosilicates [Y<sub>2</sub>SiO<sub>5</sub> (YSO) and Gd<sub>2</sub>SiO<sub>5</sub> (GSO)] are used as laser crystal hosts for doping with trivalent lasing rare-earth ions such as Dy<sup>3+</sup>, Nd<sup>3+</sup>, Yb<sup>3+</sup>, Er<sup>3+</sup>, Tm<sup>3+</sup>. Mixed (Gd<sub>1-x</sub>Y<sub>x</sub>)<sub>2</sub>SiO<sub>5</sub> (GYSO) crystals have been introduced to eliminate cleavage of the GSO and to combine the benefits of the ground state splitting of

the GSO with good mechanical properties of YSO (Nd<sup>3+</sup>:GYSO, Yb<sup>3+</sup>:GYSO). A continuous tenability from 1004 to 1110 nm has been realized for a continuous-wave Yb<sup>3+</sup>:GYSO laser. A passively mode-locked Nd<sup>3+</sup>:GYSO laser has been demonstrated using a SESAM with the pulse width of as short as 5 ps. Using Yb:GYSO crystals, mode-locked laser pulses

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